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SAMPLE POROSITY ON X-RAY INTEGRATED INTENSITIES

REPORT NO. 114 (Revised)
5010.11.842
DA 593-32-007

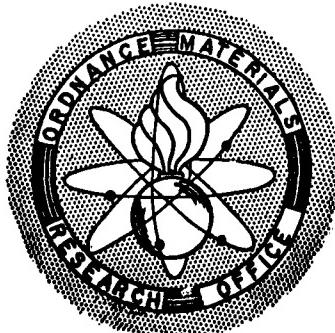
J. J. DeMARCO
and
R. J. WEISS

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Authors:


J. J. DEMARCO
Physicist
Materials Research Laboratory


R. J. WEISS
Physicist
Materials Research Laboratory

Approved:


R. BEEUWKES, JR.
Chief Scientist
Ordnance Materials Research Office

January 1963

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ORDNANCE MATERIALS RESEARCH OFFICE
WATERTOWN ARSENAL
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**AN EXPERIMENTAL DETERMINATION OF THE EFFECT OF POWDER
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J. J. DeMARCO and R. J. WEISS

Materials Research Laboratory
Ordnance Materials Research Office
Watertown Arsenal
Watertown 72, Massachusetts

ABSTRACT

Some measurements are presented which support a simple derivation for the effect of porosity on the integrated intensity of Bragg peaks in powders. A practical method of correcting for porosity is presented which makes use of fluorescent radiation from the sample.

AN EXPERIMENTAL DETERMINATION OF THE EFFECT OF POWDER SAMPLE POROSITY ON X-RAY INTEGRATED INTENSITIES

J. J. DeMARCO and R. J. WEISS

It is the purpose of this note to underscore the fact that porosity (or surface roughness) can give rise to large errors in X-ray intensity measurements on powders and to propose a simple semi-empirical expression for estimating the effect of porosity and a practical method for correcting for it.

If one is measuring the Bragg peaks from an infinitely thick powder sample in symmetrical reflection then the integrated intensity in absence of extinction is ordinarily taken to be proportional to $Q/2\mu$ (James 1954, p. 51) where μ is the linear absorption coefficient of the bulk material and Q is the integrated reflecting power per unit volume of a single crystal-lite. This expression is valid either for a powder sample whose apparent density equals the true density of the bulk material or for a powder sample whose particle size is so small that the X-ray traverses a great many particles. However even for a particle size as small as a few microns the absorption coefficient for elements of $Z > 25$ is so high as to limit the depth of penetration of X-rays to only a few particles. Thus we can expect that the X-rays will traverse a statistical distribution of path lengths on entering and leaving a typical powder sample. On the average, though, the emergent path lengths are greater than the path lengths on entering. Since the Bragg intensity is proportional to the amount of sample traversed by the X-rays and the absorption of X-rays is exponential there is a different averaging for the two competing processes. It turns out that this reduces the intensity from that expected from $Q/2\mu$ by as much as 20% in typical cases encountered in X-ray diffraction.

We have calculated the integrated intensity in infinitely thick powder samples in symmetrical reflection on the simple assumption that the X-rays

encounter a distribution in emergent path lengths $\exp[-\epsilon|\Delta|n/(1-\alpha)]$ where n is the average number of particles traversed by the X-ray, $1 + \Delta$ the correction to the emergent path length, ϵ an adjustable parameter and α the ratio of the average path length in the powder sample to the path length in a bulk sample, i.e., the ratio of the apparent density of the powder to the true density of the solid. As is expected such a distribution function becomes narrower as either the apparent density or n increases and becomes a delta function when $\alpha = 1$. The reason for the longer emergent path lengths can be seen in Fig. 1 where an entering X-ray is seen to penetrate quite deeply due to voids and thus cannot easily leave. If the reverse ray were equally intense (reverse the arrows in Fig. 1), there would be no porosity effect but such a reversed ray would have been scattered or absorbed before reaching this depth.

If we let d equal the average particle size then $n \approx 1/\mu d$ and a straightforward integration yields (x is the depth from the surface)

$$I \propto Q \int_0^\infty dx \int_r^\infty \frac{\alpha}{\sin\theta} \exp\left[\frac{-2\mu ax(1+\Delta)}{\sin\theta}\right] \exp\left[\frac{-\epsilon n |\Delta|}{1-\alpha}\right] d\Delta / \int_0^\infty \exp\left[\frac{-\epsilon n |\Delta|}{1-\alpha}\right] d\Delta$$

$$I \propto \frac{Q}{2\mu} \cdot 1 - \left[\frac{(1-\alpha)\mu d}{2\epsilon} \right] \quad (1)$$

valid for $\mu d < 1$

showing that the intensity decreases from that expected by the bulk sample, $Q/2\mu$. In the limit as either μ or d approaches zero or α approaches unity the intensity for the bulk sample is realized.

In Fig. 2 are the results of integrated intensity measurements of the 111 and 200 reflections taken from ten micron size aluminum powder compressed into briquets under varying pressures so as to vary the density from 1.28 gm/cm³ to 1.95 gm/cm³ (true density 2.70 gm/cm³). At MoK α ($\mu = 13.5/\text{cm}$)

the X-rays penetrate about 100 particles and the intensity of the (111) peak appears independent of packing density. At Cr K α ($\mu = 400/\text{cm}$) the X-rays only penetrate a few particles and both the (111) and (200) peaks show intensity loss in the low density samples. The best fit of eq. 1 (solid lines in Fig. 2) is obtained by adjusting the parameter ϵ so that $\epsilon \approx 1.0$.

Using this value of the parameter ϵ , Fig. 3 shows the results of comparing eq. 1 with intensity measurements of the 110 reflection taken from five micron carbonyl iron powder at Ag K α ($\mu = 143/\text{cm}$), Mo K α ($\mu = 295/\text{cm}$), Cr K α ($\mu = 906/\text{cm}$) and Cu K α ($\mu = 2500/\text{cm}$). For Ag K α the X-rays penetrate about fifteen particles while for Cu K α only one to two particles are penetrated. After making a small correction for preferred orientation in the high density samples, the good agreement with eq. 1 (solid lines) over a wide range of density and absorption coefficients leads us to believe that eq. 1 is useful in estimating the porosity effect.

If the sample is caused to fluoresce by the incident radiation then a measurement of the fluorescent intensity relative to the fluorescent intensity from a bulk polished sample enables one to make a direct estimate of the effect of porosity on the Bragg intensity without requiring a knowledge of a or d . Employing the same formulation that led to eq. 1 and replacing μ by $(\mu + \mu^*)/2$ we find that the integrated intensity of the Bragg peak is reduced by a factor R given by

$$R \approx 1 - \frac{(\mu + \mu^*)(1 - R^*)}{2\mu^*} \quad (2)$$

which is independent of the distribution function selected and where R^* is the ratio of the fluorescent intensity from the porous sample to that from a polished bulk sample, μ the linear absorption coefficient of the incident radiation and μ^* the average linear absorption coefficient of the fluorescent radiation (FeK α and FeK β in our measurement).

In Fig. 4 the measured values of R for Cu $K\alpha$ (Fig. 3) are plotted against the right side of eq. 2 utilizing measured values of R^* . The agreement is good to $< 10\%$ and leads us to suggest the use of eq. 2 as a practical method for estimating the correction for porosity, particularly when the correction is small.

There have been previous considerations of the effect of porosity (Taylor (1944), Brentano (1935), Schafer (1933), Brindley (1945), Rusterholz (1931), de Wolff (1947), Wilchinsky (1951). Several of the authors have given elaborate derivations which in the main agree with eq. 1 (except for some differences in the power of the term $(1-\alpha)$ depending on the model) but none of the expressions yield any better representation of the data in Figs. 2 and 3 than our simply derived expression. (An examination of powder particles under a microscope indicates that their irregular shapes are hardly amenable to a sophisticated calculation). Wilchinsky has made some measurements on powders of particle size $0.3 < \mu d < 16$ principally to show that a treatment such as in eq. 1 is not valid for $\mu d \gg 1$.[†] Batterman, Chipman and DeMarco (1961) have utilized the fluorescent intensity of their compressed powder samples to eliminate porosity effects by selecting a sample whose fluorescent intensity equalled that of a solid specimen. Figure 4 and eq. 2 lend support to their procedure.

[†]In such a case it may be reasonable to replace μd by μd^* where d^* is an effective depth of penetration into the particle. This should not affect eq. 2, though, since it is independent of the distribution function.

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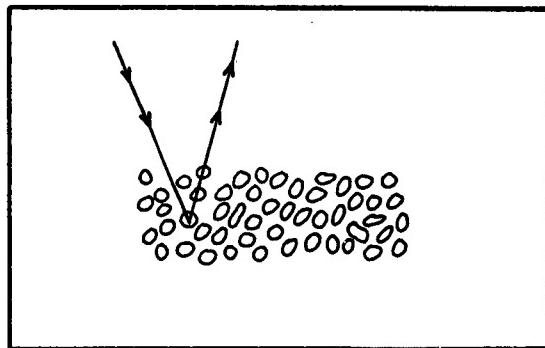


Fig. 1. A schematic sketch showing that, on the average, emergent path lengths in the sample are greater than entering path lengths since a certain fraction of X-rays can enter quite deeply through voids.

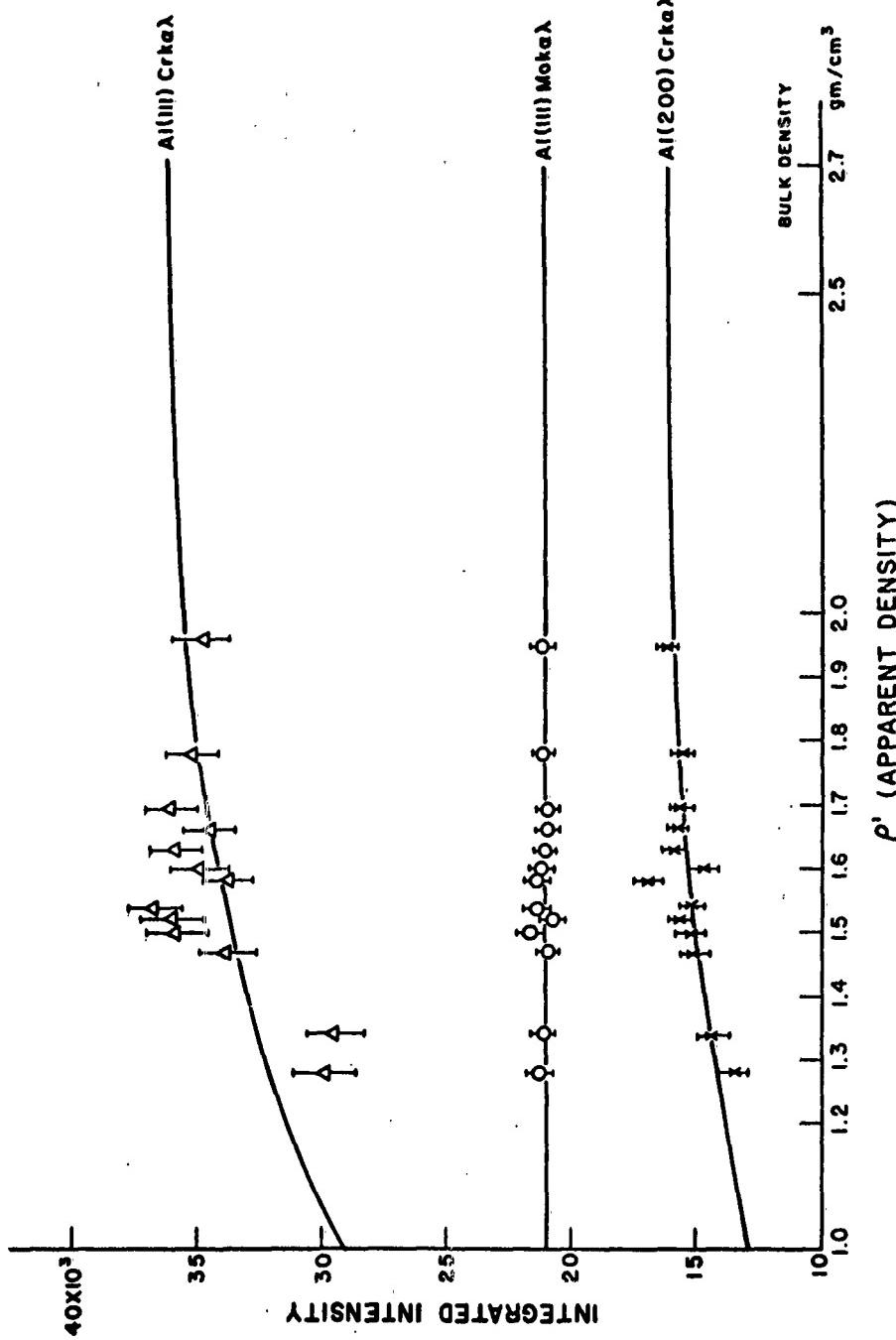


Fig. 2. The integrated intensity of the Al 111 peak at Mo $\kappa\lambda$ and the Al 111 and 200 peaks at Cr $\kappa\lambda$ as a function of the apparent density of the powder sample. The solid curves are an empirical fit of eq. 1 for $\epsilon \approx 1.0$ and d (measured) $\approx 10^{-3} \text{ cm}^3$.

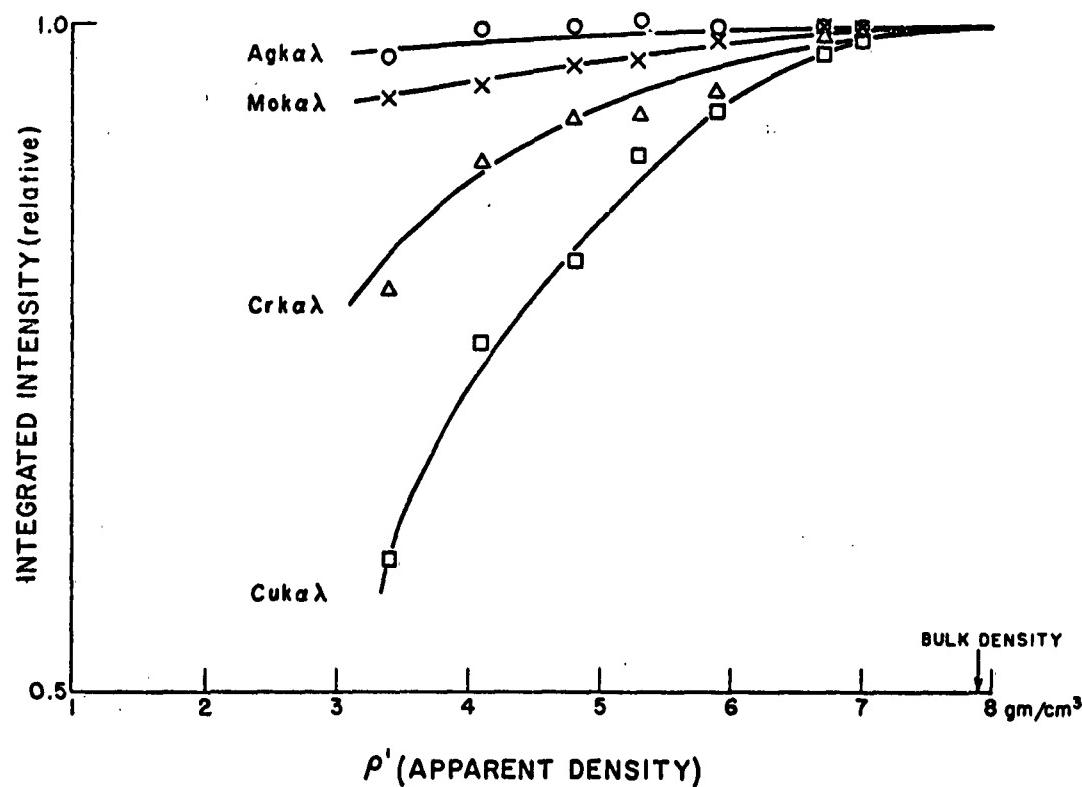


Fig. 3. The integrated intensity of the Fe 110 peak at Ag $\text{K}\alpha$, Mo $\text{K}\alpha$, Cr $\text{K}\alpha$ and Cu $\text{K}\alpha$ wave lengths as a function of apparent density of the 5 micron carbonyl Fe powder samples. The solid curves are determined from eq. 1 ($\epsilon = 1.0$).

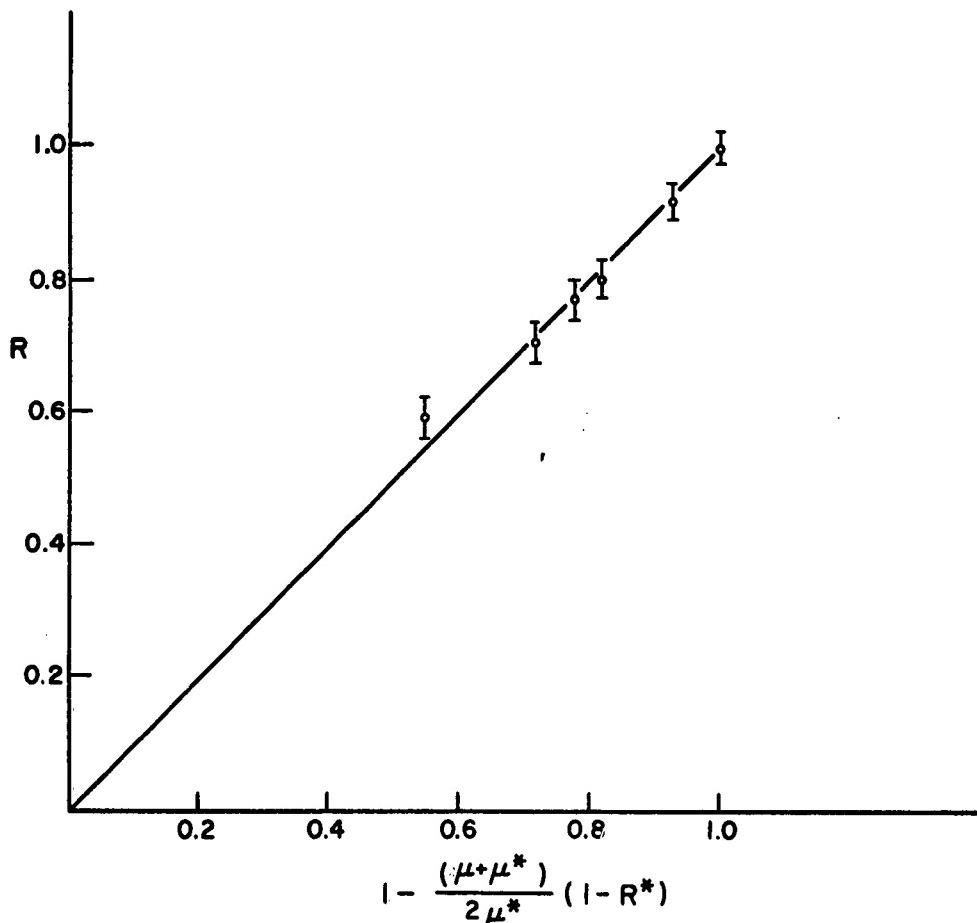


Fig. 4. The ratio of the integrated intensity of the 110 Fe peak relative to a powder sample with no porosity (extrapolated) versus the argument in eq. 2, taken with Cu K α . R^* is the ratio of the fluorescent intensity of the powder sample to the fluorescent intensity of a solid polished sample. If the fluorescent intensity of a powder sample equals the fluorescent intensity of a solid specimen of the same material then porosity effects are absent.

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